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Cooper pair formation dynamics in $Bi_2Sr_2CaCu_2O_{8+\delta}$

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Abstract: We utilize ultrafast terahertz pulses to monitor the carrier dynamics in the high-T_C superconductor $Bi_2Sr_2CaCu_2O_{8+\delta}$. The temperature, density and time dependence distinctly exposes a bimolecular recombination process of quasiparticles which underlies formation of Cooper pairs.
OCIS codes: 320.7130 Ultrafast processes in condensed matter, 300.6340 Spectroscopy, infrared

Understanding the elusive mechanism of high-T_C superconductivity in the strongly-correlated cuprates requires a microscopic picture of quasiparticle interactions underlying the formation of Cooper pairs. The potential of ultrafast spectroscopy to reveal carrier interactions motivated time-domain experiments on cuprates, many of them probing the dynamics at energies

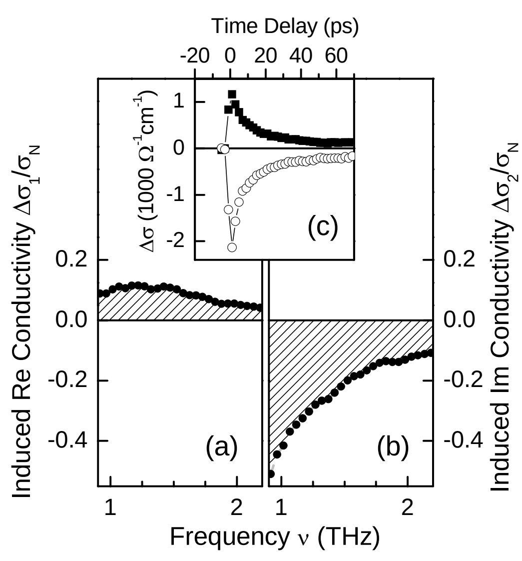

Fig. 1. Induced changes in the optical conductivity of optimally-doped Bi-2212 (transition temperature $T_c \approx 88$ K, 62-nm thickness). (a,b) spectral changes normalized to the normal state conductivity σ_N directly after excitation $\Delta t \approx 1$ ps, for $T = 5$ K and absorbed excitation fluence I_0 = 17 nJ/cm^2 per CuO_2 bilayer. (c) time evolution of transient conductivity at a representative probe frequency 1.3 THz and excitation fluence $I = I_0$ (squares: real part, open circles: imaginary part).

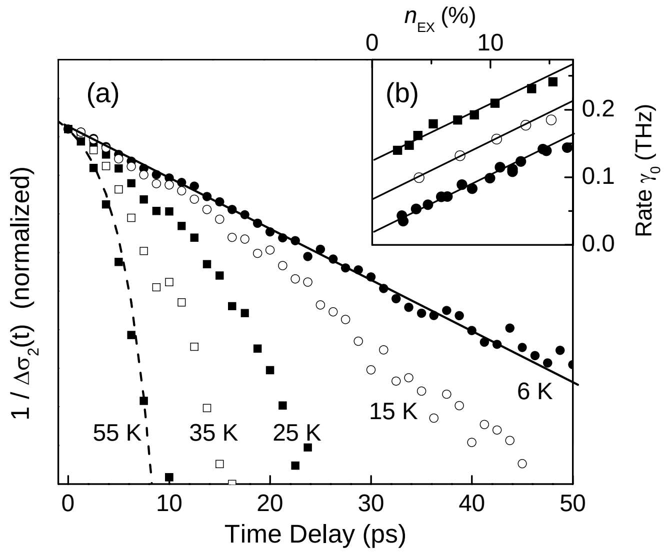

Fig. 2. (a) Conductivity dynamics plotted on a inverted ordinate $1/\Delta\sigma_2(t)$, for different temperatures, $I \approx I_0$, and probe frequency 1.3 THz. Bimolecular (solid line) and exponential (dashed line) kinetics are shown for comparison. (b) Initial decay rate γ_0 versus photoinduced condensate depletion at $T = 5$ K (dots), 20 K (open circles), 30 K (squares).

E ~ 1.5 eV far above the superconducting gap [1]. Only recently, first experiments emerged that detect the low-energy response, providing in turn direct access to pseudogap, quasiparticle, and Cooper-pair responses [2,3]. Here, we report the first study probing ultrafast THz conductivity changes in the high-T_C superconductor $Bi_2Sr_2CaCu_2O_{8+\delta}$ (Bi-2212). The measurements reveal a bimolecular formation kinetics of Cooper pairs on a picosecond timescale, yielding an otherwise hidden measure of quasiparticle recombination.

We investigate c-axis oriented Bi-2212 films grown via molecular beam epitaxy. Near-infrared femtosecond pulses from a 250-kHz Ti:sapphire amplifier excite the films, and the low-energy response is probed with temporally delayed picosecond THz pulses. Detection of the transmitted THz field and its transient change via electro-optic sampling yields both the equilibrium CuO_2-plane optical conductivity $\sigma(\omega)$ and its pump-induced transient change $\Delta\sigma(\omega)$. The real part σ_1 provides a measure of quasiparticle densities, while the imaginary part σ_2 is governed by the inductive $1/\omega$ response of the condensate. Fig. 1 shows induced changes that occur after optical excitation in the superconducting state. Directly after excitation ($\Delta t \approx 1$ ps), a strongly frequency-dependent reduction $\Delta\sigma_2(\omega)$ of the imaginary part is observed (Fig. 1b) along with a marked increase in the real part (Fig. 1a). These features substantiate a depletion of the superconducting condensate accompanied by transfer into excess quasiparticles. The simultaneous decay of the induced changes in real and imaginary parts (Fig. 1c) marks the picosecond return of quasiparticles into the superconducting condensate.

The kinetics is strongly non-exponential at low temperatures, but plotting the inverse $1/\Delta\sigma_2(t)$ we find a strikingly simple linear time dependence (T=6 K, Fig 2a). This dependence is a well-known hallmark of bimolecular kinetics. With $\Delta\sigma(t)$ proportional to the excess quasiparticle density $n_{EX}(t)$, it implies a dynamics $d/dt\, n_{EX}(t) = -\beta \cdot n_{EX}(t)^2$ where quasiparticles recombine into Cooper pairs via pairwise interaction. We obtain a recombination coefficient $\beta \approx 0.05$ cm^2/s which can be further compared with theory [4]. At higher temperatures, interactions of photoinduced quasiparticles with thermally-excited quasiparticles n_{th} are relevant, and an additional term $2\beta n_{EX}(t) n_{th}$ adds to the kinetics which for $n_{th} \gg n_{EX}$ implies a single-exponential decay. Indeed, Fig. 2(a) shows that the dynamics evolves from *t*-linear bimolecular kinetics ($T = 6$ K) to exponential decay at higher temperatures.

The mixed character at intermediate temperatures can be accessed through the instantaneous initial decay rate $\gamma(0) = n_{EX}^{-1}\, d/dt\, n_{EX}(t)|_{t=0} = -\beta \cdot n_{EX} + 2\beta \cdot n_{th}$. This linear dependence on excitation density is experimentally observed at sufficiently low temperatures and excitation densities (Fig 2b). Extrapolation to vanishing excess density gives the equilibrium recombination rate which is far smaller than the ≈2 THz momentum scattering rates. This underlines the distinctly different physical nature between inelastic recombination events connected to Cooper pair formation and elastic quasiparticle scattering with large momentum transfer responsible for transport properties. Our experiment thus provides strong evidence that the temporal evolution of the THz conductivity reflects a quasiparticle-quasiparticle scattering process that underlies Cooper pair formation. We will also discuss the behavior for underdoped and overdoped Bi-2212 and excitation at lower photon energies.

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[1] N. Gedik, J. Orenstein, R. Liang, D. A. Bonn, and W. N. Hardy. "Diffusion of Nonequilibrium Quasi-Particles in a Cuprate Superconductor," Science **300**, 1410–1412 (2003), and references therein.
[2] R. A. Kaindl *et. al.* "Ultrafast mid-infrared response of $YBa_2Cu_3O_{7-\delta}$," Science **287**, 470–473 (2000).
[3] R. D. Averitt *et al.*, "Nonequilibrium superconductivity and quasiparticle dynamics in $YBa_2Cu_3O_{7-\delta}$," Phys. Rev. B**63**, 140502 (2001).
[4] J.-J. Chang, and D. J. Scalapino, "Kinetic-equation approach to nonequilibrium superconductivity," Phys. Rev. B**15**, 2651–2670 (1977).